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N_2O as an indicator of Arctic vortex dynamics: Correlations with O_3 over Thule, Greenland in February and March, 1992

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Abstract. We have recovered vertical profiles of stratospheric N_2O from spectra observed using a ground-based mm-wave spectrometer during the Arctic spring. The measurements were made from Thule, Greenland (76.3°N, 68.4°W) on nine occasions from late February to late March, 1992 as part of the Upper Atmosphere Research Satellite (UARS) Correlative Measurements Program and the European Arctic Stratospheric Ozone Experiment (EASOE). During late February Thule was under the inside edge of the Arctic vortex and mixing ratio profiles measured in that period are substantially reduced from typical high-latitude summer values. By late March the polar vortex had moved well away from Thule and N_2O mixing ratios were greatly increased, coinciding with a basic change in circulation that brought in air from the Aleutian high. The motion of the vortex is also illustrated in the change in potential vorticity above Thule. A correlation with ozone balloonsonde data from Thule is made and compared to similar analyses of the Airborne Arctic Stratospheric Expedition (AASE) measurements. Within the sensitivity of our analysis, the correlation of N_2O and O_3 shows no evidence of ozone depletion within the vortex during this period; however, there is a distinct difference in the correlation inside and outside the vortex.

The Arctic polar vortex is much more variable and limited in size than the Antarctic vortex, and in order to understand better the chemical changes in the polar stratosphere, it is essential to understand the dynamical influences. Nitrous oxide is a good dynamical tracer in the stratosphere. Its only sources are at ground level and its lifetime in the lower stratosphere is on the order of 100 years. Following the ground-based measurements in Antarctica in 1986 by Parrish et al., [1988], intensive aircraft and balloon measurements of N_2O have been made in the polar winter vortex regions [*e.g.,* Lowenstein et al., 1990]. Ground-based measurements, however, offer advantages of greater vertical range and more frequent monitoring.

We have observed N_2O spectra on nine days between February 22 and March 25 using a mm-wave heterodyne receiver with a superconducting tunnel junction mixer and filter bank spectrometer [de Zafra et al., 1992]. We measure the pressure-broadened emission intensity of the N_2O molecular rotation line at 276.328 GHz (1.08 mm). Each spectrum is the result of $\approx$ 2 hours integration near midnight. The spectrometer has 512 channels, each of width 1 MHz, allowing retrieval of vertical profiles between approximately 16 and 50 km. The pressure-broadening coefficient of N_2O is 2.38 MHz/mb [Lacome et al., 1984], so a Lorentzian-shaped line contribution from an altitude of 16 km will have a half-width at half-maximum of $\approx$ 250 MHz. Broader lines (emitted from lower altitude) contribute diminishing information within our bandpass limits, for purposes of vertical profile recovery, and appear increasingly as constant offsets from zero intensity. Lines narrower than 1 MHz are unresolved, so the center channel contains the integrated intensity above $\approx$ 50 km, which in any case is negligible for N_2O. The mixing ratio as a function of altitude is determined by numerical deconvolution of the spectra. We have used an iterative method based on a modified Chahine-Twomey inversion algorithm [Twomey et al., 1977]. The resulting profiles have a vertical resolution of roughly one scale height [see *e.g.,* Bevilacqua and Olivero, 1988], which in the Arctic is approximately 7 km. Because our data-taking technique requires observation at high zenith angles through a large airmass, the instrument always faced north and detected radiation emitted from $\approx$ 100 km away. Spectra presented here are scaled to equivalent zenith intensity, thus the recovered profiles are *vertical* distributions at the mean location of the source.

For the period of our measurements the polar vortex was usually centered over the Barents Sea, so Thule never saw air from the core of the vortex, but only from the inside edge, and later in March from the exterior. To define the edge of the vortex, the steepest gradient of potential vorticity (PV) was used. From the maps produced by the European Centre for Medium-range Weather Forecasts and the Norwegian Institute for Air Research (NILU) for EASOE, we have approximated the steepest gradient as coinciding with PV of $78 \times 10^{-6}\mathrm{Km^2/kgs}$ for the 550K potential temperature (θ) level ($\approx$ 22 km). Back

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trajectories, calculated by B. Knudsen and made available on NILU's Atmospheric Database for Interactive Retrieval (NADIR), have also been used to determine the source of the air observed at Thule. During late February Thule was inside the vortex, 400 to 1000 km from the edge, and back trajectories indicate the air flow was from the north along the inside edge of the vortex. During March 6 – 18 the vortex gradually shifted so that Thule was just outside the edge, although back trajectories for θ = 550K show that air flow was still associated with the vortex (see further discussion below). After March 18 Thule was more than 2000 km from the vortex edge, and air above Thule had shifted to a flow associated with the Aleutian high-pressure anti-cyclonic center. Our measurements on February 22, 24, 26, 28 and March 1 all give similar vertical profiles showing low mixing ratios much like those from the Antarctic spring, now established as indicating subsidence of the air within the vortex [*e.g.*, Parrish et al., 1988; Proffitt et al., 1990]. The data from March 9, 10, 23, 25 are more variable, but all have higher mixing ratios at all altitudes than the data from earlier in the spring, particularly between 24 and 40 km.

The scaled spectral data for two days are shown in Figure 1, where the emission line intensity, given as equivalent

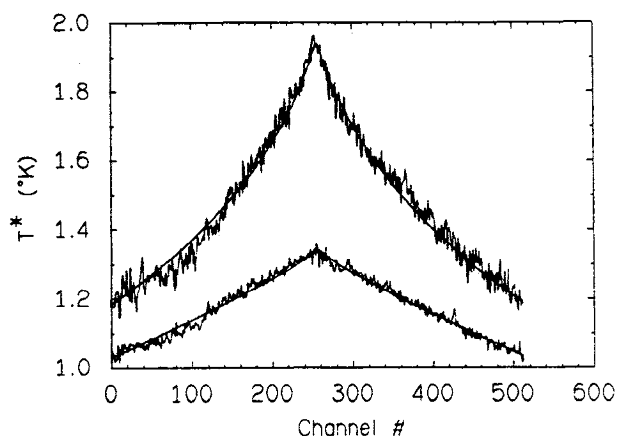

Fig. 1. The observed spectra from February 24 (lower curve) and March 23 (upper curve) with deconvolution fits. Intensity, as equivalent black-body radiation in Kelvin, is plotted versus relative frequency in MHz. Channel 256 corresponds to 276.328 GHz.

black-body radiation temperature, is plotted as a function of spectrometer channel (1 MHz/channel). February 24 and March 23 were chosen as representative days when Thule lay inside and outside the vortex, respectively. Vertical profiles, obtained by deconvolution of these spectra, are shown in Figure 2. Spectra synthesized from the derived profiles are overlaid on the measured spectra in Figure 1. A constant mixing ratio of 307 ppbv throughout the troposphere is assumed [Weiss, 1981 and Loewenstein et al., 1990]. Although we cannot determine the mixing ratio below ≈ 16 km, the profiles are shown from 10 km, assuming a smooth decrease from the tropospheric value. The shaded regions around the two profiles show the range of profiles giving spectra that match our data within the noise and also include the uncertainties in calibration.

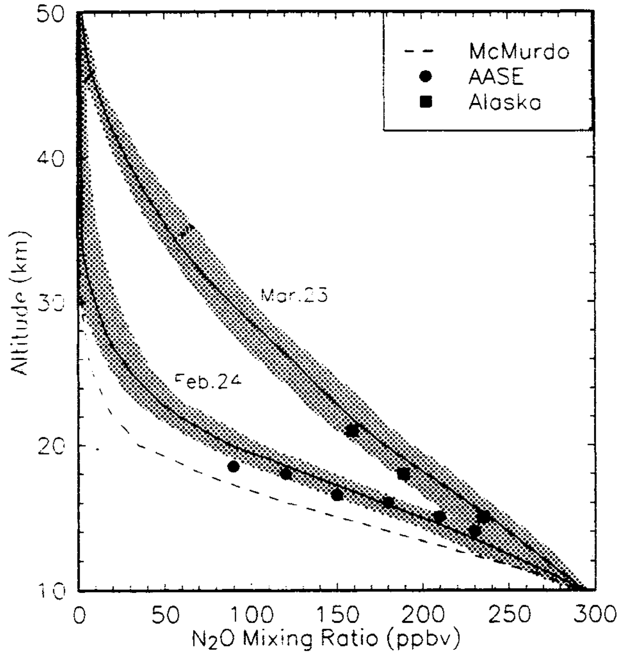

Fig. 2. Vertical profiles from deconvolution of the spectra in Figure 1 (solid curves) with measurement and analysis uncertainties (shaded regions). The dashed curve is a mm-wave spectroscopy result from McMurdo, Antarctica, September, 1986 [Parrish et al., 1988]. AASE 1989 results are averages of 8 flights more than 2 degrees latitude inside the vortex, February, 1989 [Loewenstein et al., 1990]. Aircraft results from Alaska, June, 1977 [Vedder et al., 1981] have uncertainties of ±15%.

Other measurements of polar N_2O are included in Figure 2 for comparison. The dashed curve is from mm-wave spectroscopic measurements at McMurdo Station, Antarctica in September, 1986 [Parrish et al., 1988]. The profile from February 24 is higher than the McMurdo data, indicating less subsidence than in the Antarctic. The AASE results shown are an average from eight flights more than 200 km inside the vortex in February, 1989 [Loewenstein et al., 1990]. The Alaska results are high-altitude aircraft measurements in June, 1977 at 73°N [Vedder et al., 1981]. The aircraft measurements cover smaller ranges of altitude, but agree quite well with our profiles.

Figure 3 shows all of our derived mixing ratio profiles. As the vortex moved away from Thule, given mixing ratio values moved to higher altitudes, coinciding with a decrease in potential vorticity (Figure 4) and an increase in temperature. The profile from March 9 is particularly interesting in that it shows that the transition from inside

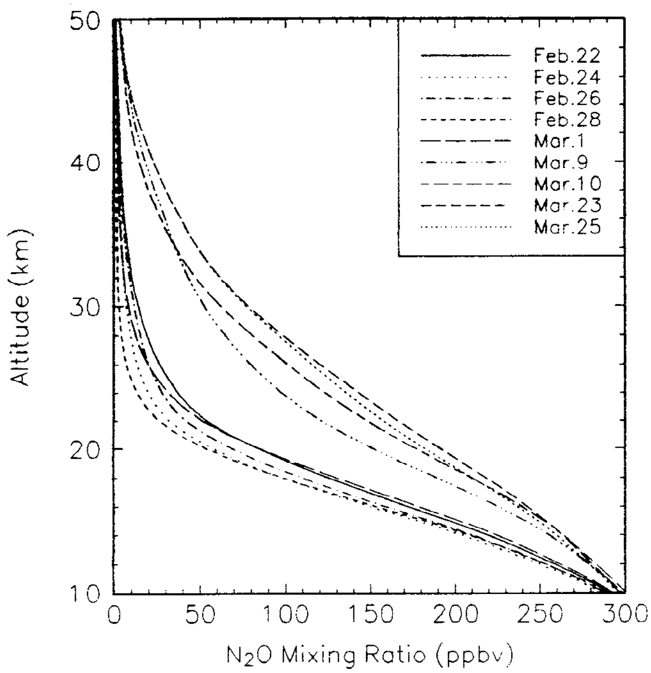

Fig. 3. Vertical profiles for each spectrum observed at Thule, 1992. Uncertainties for each curve are similar to shaded regions shown in Figure 2.

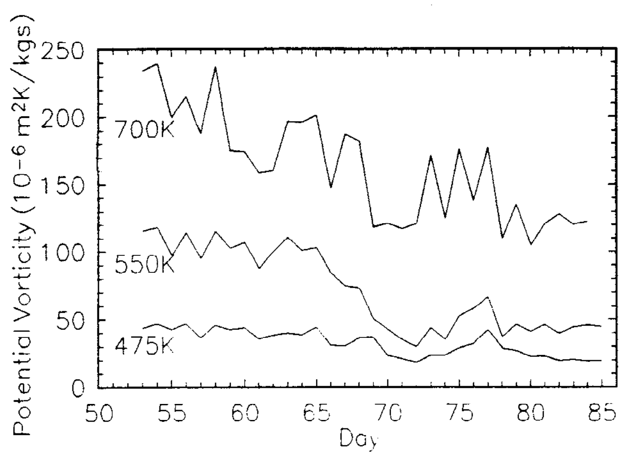

Fig. 4. Potential vorticity for three potential temperature levels over Thule for the period of our measurements (day 53 = February 22 to day 86 = March 25). θ = 475K corresponds to $\approx$ 19 km, 550K to $\approx$ 22 km, 700K to $\approx$ 25 km.

to outside the vortex did not occur simultaneously at all altitudes. There is an increase in mixing ratio at high altitudes, but little change at low altitudes, indicating that the air at high altitudes was coming from the Aleutian high, while at low altitudes the flow was still from the vortex. Figure 4 also illustrates this feature; potential vorticity on the θ = 700K and θ = 550K levels drops on March 9, but not until the following day on the θ = 475K level.

The correlation of N_2O mixing ratios with O_3 has been shown to be a useful method for studying the amount and location of ozone depletion in both the Antarctic and Arctic vortex regions [Strahan et al., 1989, Proffitt et al., 1990, Proffitt et al., 1992]. Under "normal" conditions in the lower stratosphere N_2O decreases with altitude while O_3 increases, so the correlation is negative. This relationship will persist during vortex subsidence. When ozone is chemically depleted, however, the correlation changes, and can even become positive for severe depletion. Ozonesondes were launched regularly from Thule during February and March, and we have been able to make a similar correlation for 6 days when both N_2O and O_3 were measured less than 12 hours apart. In Figure 5 ozone mixing ratio is plotted versus N_2O mixing ratio with altitude as an implicit variable. The altitude range starts at 10 km in the lower right, running to the limit of the balloon data, typically $\approx$ 30 km. For comparison the relationship derived for the 1989 AASE aircraft data is included on the plot. The solid line between diamonds is the "Exterior

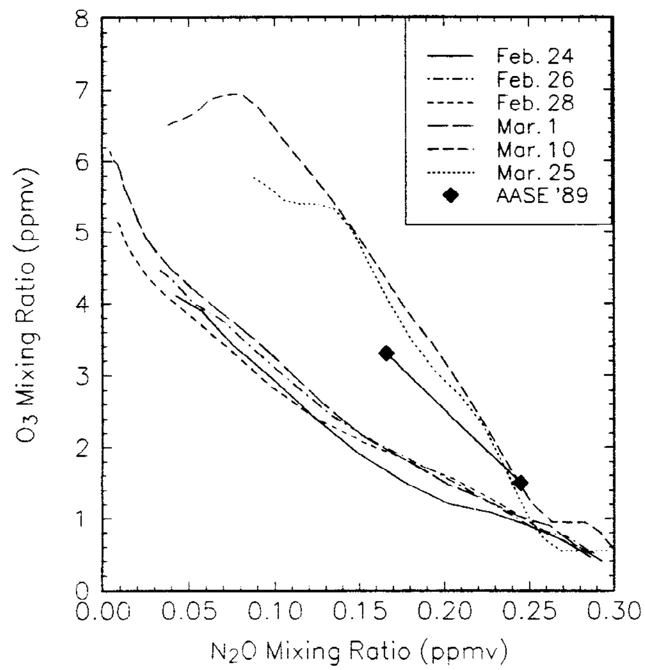

Fig. 5. Correlation of N_2O and O_3. Our derived vertical profiles are plotted against ozonesonde data taken from Thule. Altitude range on each day is 10 km to $\approx$ 30 km. The solid line between diamonds is the "Exterior Fit" to AASE 1989 data for correlations outside the vortex [Figure 1d, Proffitt et al., 1992].

Fit" from Figure 1d of Proffitt et al. [1992] for the range of their data. The slope change in curves at high ozone values for March 10 and 25 is due to passage of the balloons through the ozone mixing ratio maximum at about 30 km, and does not indicate chemical depletion.

The curves for March 10 and 25 in Figure 5 are distinctly different from the results from inside the vortex.

The data from inside the vortex has a similar slope to the aircraft "Exterior Fit", but is somewhat offset. This difference may be attributable to the differences in time and location of the measurements: the aircraft flights took place during January and early February, 1989, near Scandanavia. None of our curves for the "inside" data show any statistically significant change in slope with decreasing N_2O concentration, which would indicate chemical ozone depletion. This agrees well with the lack of large concentrations of ClO measured at the same time [de Zafra et al., this issue]. Our late March results show that air originating from the Aleutian high has a very different N_2O-O_3 relationship compared to the other results shown here: there is more ozone for given amounts of N_2O. There are typically high amounts of ozone associated with the Aleutian high, and Figure 5 demonstrates that there were not proportionately high amounts of nitrous oxide.

Our measurements of the vertical profile of nitrous oxide from late February through March show a large change in mixing ratio as the polar vortex moved away from Thule. These results have a greater vertical range than previous data for the Arctic region and show that the vortex edge can be quite variable with height. Within the sensitivity of these measurements, correlation of ozone and nitrous oxide shows no indication of ozone depletion over Thule during this time.

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References

Bevilacqua, R.M. and J.J. Olivero, Vertical resolution of middle atmospheric measurements by ground-based microwave radiometry, *J. Geophys. Res., 93,* 9463, 1988.

de Zafra, R.L., W.H. Mallison, M. Jaramillo, J.M. Reeves, L.K. Emmons, and D.T. Shindell, A new high-sensitivity superconducting receiver for mm-wave remote-sensing spectroscopy of the stratosphere, *Proceedings of the Quadrennial Ozone Symposium,* in press, 1992.

de Zafra, R.L., L.K. Emmons, J.M. Reeves, and D.T. Shindell, An overview of millimeter-wave spectroscopic measurements of chlorine monoxide at Thule, Greenland, February-March, 1992: Vertical profiles, diurnal variation, and long-term trends, *this issue.*

Knudsen, B., P. von der Gathen, G.O. Braathen, R. Fabian, T.S. Jørgenson, E. Kyro, R. Neuber, and M. Rummukainen, Temporal development of the correlation between ozone and potential vorticity in the Arctic in the winters of 1988/1989, 1989/90, and 1990/91, *Proceedings of the Quadrennial Ozone Symposium,* in press, 1992.

Lacome, N., A. Levy, and G. Guelachvili, Fourier transform measurement of self-, N_2-, and O_2-broadening of N_2O lines: temperature dependence of linewidths, *Appl. Optics, 23,* 425, 1984.

Loewenstein, M., J.R. Podolske, K.R. Chan, and S.E. Strahan, N_2O as a dynamical tracer in the Arctic vortex, *Geophys. Res. Lett., 17,* 477, 1990.

Parrish, A., R.L. de Zafra, M. Jaramillo, B. Connor, P.M. Solomon, and J.W. Barrett, Extremely low N_2O concentrations in the springtime stratosphere at McMurdo Station, Antarctica, *Nature, 332,* 53, 1988.

Proffitt, M.H., S. Solomon, and M. Loewenstein, Comparison of 2-D model simulations of ozone and nitrous oxide at high latitudes with stratospheric measurements, *J. Geophys. Res., 97,* 939, 1992.

Proffitt, M.H, J.J. Margitan, K.K. Kelly, M. Loewenstein, J.R. Podolske and K.R. Chan, Ozone loss in the Arctic polar vortex inferred from high-altitude aircraft measurements *Nature, 347,* 31, 1990.

Strahan, S.E., M. Loewenstein, J.R. Podolske, W. Starr, M.H. Proffitt, K.K. Kelly, and K.R. Chan, Correlation of N_2O and ozone in the southern polar vortex during the Airborne Antarctic Ozone Experiment, *J. Geophys. Res., 94,* 413, 1990.

Twomey, S., B. Herman, and R. Rabinoff, An extension of the Chahine method of inverting the radiative transfer equation, *J. Atmos. Sci., 34,* 1085, 1977.

Vedder, J.F., E.C.Y. Inn, B.J. Tyson, C.A. Boitnott, and D. O'Hara, Measurements of CF_2Cl_2, $CFCl_3$, and N_2O in the lower stratosphere between 2°S and 73°N latitude, *J. Geophys. Res., 86,* 7363, 1981.

Weiss, R.F., The temporal and spatial distribution of tropospheric nitrous oxide, *J. Geophys. Res., 86,* 7185, 1981.

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